

## RESEARCH PAPER RP1283

Part of *Journal of Research of the National Bureau of Standards*, Volume 24,  
March 1940

## MEASUREMENT, IN ROENTGENS, OF THE GAMMA RADIATION FROM RADIUM BY THE FREE-AIR IONIZATION CHAMBER

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### ABSTRACT

The free-air ionization chamber is the accepted standard for calibrating X-ray dosage measuring instruments. With the advent of supervoltage X-rays, the need for proved standards for this range has arisen.

The present paper describes an investigation of the applicability of the free-air pressure ionization chamber for measuring radiation at the high-frequency end of the X-ray spectrum, that of the gamma radiation from radium.

Measurements were made with a narrow collimated beam of gamma rays at pressures up to 10 atm. Because of inability to obtain full current-voltage saturation, application was made of the extrapolation of reciprocal current-voltage curves in accordance with the Jaffe-Zanstra theory. This was first tested carefully with 350-kv X-rays at 1-, 6-, and 10-atm pressure and found to hold within our experimental limits. It was found that at 10-atm pressure a plate separation of about 35 cm is necessary to utilize the full electron range. Ranges in the forward direction were not effectively greater than 70 cm. Measurements of the gamma-ray emission from radium gave a value of 8.16 roentgens per milligram per hour at a distance of 1 cm, which is in line with several similar determinations by other methods.

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### I. INTRODUCTION

The free-air<sup>1</sup> ionization chamber is the accepted standard for calibrating X-ray dosage-measuring instruments. With the advent of million-volt X-rays, the need for proved standards for this range has

<sup>1</sup> A "free-air" ionization chamber is one wherein the volume from which the ions are effectively measured is surrounded by an envelope of air having a thickness in all directions at least as great as the maximum range of the secondary electrons entering or leaving the volume in those directions. For low voltages the ionization chamber is usually used with air at atmospheric pressure and is frequently termed an "open-air" ionization chamber. In this paper the use of the term "free air" is extended to those chambers operating with air at elevated pressures.

arisen. In a recent publication [1]<sup>2</sup> the application of the free-air ionization chamber, with certain pertinent modifications in technique and necessary precautions, was described, in which was proved the adequacy of the free-air chamber for measuring X-rays up to 400-kv excitation potentials. This work still leaves its application to the higher frequency radiation untested.

Equipment is under construction to extend this investigation to 1,400 kv. In the meantime, the applicability of the methods has been investigated by studying ionization produced by gamma rays of radium. These rays comprise radiations ranging from 600 to 2,000 kv, roughly comparable to 1,500-kv X-rays. The ionization produced at unit distance from 1 mg of radium is an important physical quantity, uncertainties in which arise largely from experimental difficulties inherent in realizing the definition of the roentgen for very high frequency radiations. The measurement of X-rays and gamma rays by means of the same instrument and procedure lends greater confidence to the expression of X- and  $\gamma$ -ray dosages in a common unit—the roentgen.

The ionization produced by a narrow collimated beam of gamma rays in an air ionization chamber at pressures up to 10 atm has been measured. The investigation involved:

1. A study of the dimensions and pressures required to eliminate wall effects. This involved a study of the general ionization-chamber design with particular respect to the plate spacings and diaphragm-collector distances at various air pressures so as to insure a state of full electronic equilibrium. It also included an investigation of the diaphragming and collimating system necessary to the unambiguous measurement.

2. A determination of the correction for columnar recombination and lack of saturation. Current-voltage saturation conditions were studied, and Jaffes' theory of columnar ionization was applied to determine the saturation current.

3. A determination of the fraction of the total gamma radiation reaching the ionization chamber. This involved a review of the fundamental calibration methods for radium preparations, with relation to the measurements of the emission constant of radium in accordance with the definition of the roentgen. Included in this were corrections for the absorption of the gamma rays in air, platinum, and the radium salt itself.

Finally, the various experimental results were applied to an evaluation of the emission constants of radium in roentgens. After an analysis of the various errors involved, the results were compared with similar determinations by other methods.

The use of a relatively large quantity of radium facilitated measurements at low current sensitivities and for satisfactory distances, so that quantities involving these measurements could be evaluated with high accuracy and permit the more ready localization of any other errors.

This work was made possible through the cooperation of the National Institute of Health, which lent us, for the purpose, a specially made platinum cylinder containing about one-half gram of radium.

<sup>2</sup> Figures in brackets indicate the literature references at the end of this paper.

## II. HIGH-AIR-PRESSURE IONIZATION CHAMBER

The ionization chamber employed in this investigation has been described in detail [1] in connection with supervoltage X-ray investigations, no changes having been made for this study. Briefly, it consists of a guarded-field parallel-plate chamber (75 cm long with a maximum plate separation of about 40 cm) placed in a steel pressure tank 8 ft long and 2½ ft in diameter, suitable for operating normally up to 10 atm and, if needed, at double that pressure. Plate separation and position along the axis of the cylinder are adjustable from without through suitable airtight joints. An aluminum "window" 1.0 mm thick and 6 cm in diameter permits the effectively unobstructed entry of the radiation beam.

Air was supplied from a small compressor with intake passing through a large calcium chloride dryer, and outlet through a cylinder packed with rag waste to absorb any oil vapor or other foreign matter from the pump. Several canisters of calcium chloride dryer were also placed in the tank to remove any final traces of moisture, so that all measurements could be made under dry-air conditions.

Up to about 4 atm, the pressure was measured with a multiple-tube mercury manometer [2] to an accuracy of  $\pm 0.1$  percent. At higher pressures a Bourdon gage, calibrated with a piston gage to an accuracy of  $\pm 0.3$  percent at the particular gage points of the study, was used.

Saturation voltage was supplied from either of two sources, depending upon the magnitude of field required and the electrometer system employed. With the more sensitive vacuum-tube electrometer, very small fluctuations in the saturating voltage induced large deflections of the galvanometer. By test, it was found that fluctuations in the plate voltage exceeding  $\pm 0.01$  volt were intolerable; consequently, where plate voltages of 10,000 are used, a steadiness of 1 part in  $10^6$  is required and 1 part in  $10^7$  desirable.

For potentials up to 8,000 volts, heavy duty *B* batteries were employed. Although an individual cell is probably not steady to the required degree, the fluctuations of the 5,000 cells were statistically balanced and the resultant voltage was steady to 1 part in  $10^8$ .

Above 8,000 volts, a valve rectifier-filter was required. Stabilization of these voltages within our required limits proved to be impracticable by any of the usual methods; our requirement that the output voltage be variable was one of the main difficulties. An attempt to use the manual-compensation method described by Pallister [3] showed it to be inadequate for our needs. Indirect compensation was finally accomplished through a modification of the Swann methods, whereby a definite fraction of the fluctuating plate voltage is applied to the electrometer grid circuit in such a way as to balance out the plate-voltage variations.<sup>3</sup> By this means, plate voltages up to 25 kv were available.

Careful tests were made for any possible corona or other leakage at these high plate voltages. At atmospheric pressure, corona hissing began at about 4,000 volts and break-down occurred at about 15 kv. Up to the latter point, there was no visible corona nor did any corona occur in regions such that it could be detected with our highest electrometer sensitivities. At 10 atm, tests up to 30 kv showed no break-

<sup>3</sup> This will be described elsewhere.

down or visible corona, nor was any leakage effect detected by the electrometer. It is interesting to note that after any prolonged pumping of air into the cylinder, the electrometer showed erratic deflections attributable to stray ions (or probably ion clusters) caused by the air friction. At times it required 24 hours' rest after pumping for this effect to completely disappear. Broxon and Meredith [4] found similar disturbances in their very much smaller system and sometimes allowed several days for "aging" of the air.

Current measurements were made by essentially the same means used in our earlier X-ray measurements [1]. For comparative current measurements, the ion current was balanced against a known current through a very high (S. S. White) resistor. For absolute measurements, the same electrometer system was used as a null detector in a capacitance compensator system [5].

In both comparative and absolute measurements, the current was read in terms of a known voltage applied across the resistor or capacitor. This voltage was adjusted and read directly on an L & N type *K* potentiometer with an accuracy far beyond our requirements.

The need for shielding from radiation around the electrometer and conductors was carefully investigated. The radiation field was considerably restricted by the strict shielding and collimation at the radium as described below.

The electrometer tube was placed in a lead cylinder (wall, one-fourth in.) which in turn was supported just outside the end of the pressure tank opposite to and about 10 ft from the radium. Under these conditions, and with a relatively large testing potential difference applied between the grid circuit and the shield, no ionization current was detectable at the highest sensitivities used. In normal operation there is practically zero potential difference between grid leads and case.

In the capacity compensating system the capacitor had an ionizable volume of about 20 cm<sup>3</sup> and was subject to a potential difference of several volts between plates. This, shielded by about 2 cm of lead shot, also showed no evidence of stray ionization between plates. A third electrometer-tube housing, having no lead shielding but capable of being evacuated, was also tested and the system found insensitive to radiation either at 760-mm or at 10<sup>-2</sup>-mm pressure.

Statistical variations in ionization produced by the radium caused random fluctuations of the electrometer zero amounting to about 2 percent of the measured current when using the galvanometer employed in earlier studies. These fluctuations were reduced to a level of about 0.5 percent of the working deflection by increasing the galvanometer period, although the measurements were thereby made somewhat more laborious.

### III. DIAPHRAGMS

For most comparative measurements the construction, thickness, and alinement of the diaphragm are not important so long as the open and closed positions are reproducible. For absolute measurements, however, the thickness must be sufficient to reduce the radiation leakage through the walls of the diaphragm to less than 0.1 percent of that through the orifice. To avoid the thick diaphragms necessary with radium, most workers have collimated the radiations

by imbedding the radium in a lead block at the bottom of a deep well with walls of requisite taper. This collimating arrangement, requiring the presence of some sort of shutter system, makes it somewhat difficult to properly evaluate the scattered and background radiation. In an endeavor to avoid this difficulty, as well as to retain a system suitable for measuring X-rays of very high voltage, a diaphragm arrangement was constructed on the same general principle as for the measurement of such X-rays.

A practice in the past has been to use diaphragms which were too thin and to make corrections by subtracting the reading made with

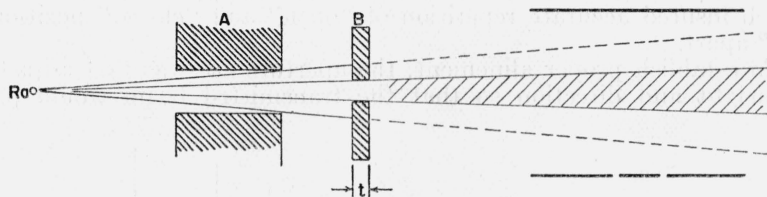


FIGURE 1.—Diagram of thin-diaphragm system.

the diaphragm closed from that made with the diaphragm open, thereby supposedly eliminating the background radiation. For radium, this procedure is not valid for the following reason: Take a gamma-ray beam partially limited by a lead block, *A* (fig. 1), and further by the limiting diaphragm, *B* of thickness *t*. With the aperture open, the total radiation measured is the unobstructed part passing through the aperture plus that which passes through the surrounding wall. With the aperture closed by a plug, the radiation measured is again that which passes through the wall plus that which

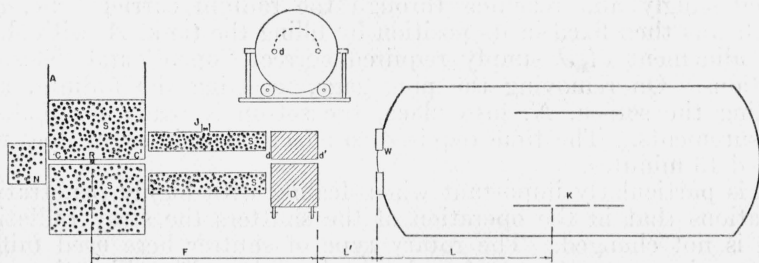


FIGURE 2.—Diagram of thick diaphragm and collimating system.

passes through the plug. The difference in the two measurements gives that passing through the aperture plus that which passes through the plug, and the result is consequently in error by the latter amount. This error is reduced by increasing the thickness *t*, of the diaphragm. Since, then, the resultant quantity is a function of the diaphragm thickness *t*, the thickness should be such that no appreciable amount of radiation passes through it.

The arrangement of the system finally adopted is shown diagrammatically in figure 2. *N* is a 5-in. lead screening cube (or box filled with 0.02-in. lead shot) mounted on rollers; *A* is an iron can (35-cm diam, 50 cm high) filled with lead shot in which is mounted, opposite appropriate windows, a thin copper tube, *cc'*, 10-mm inside diameter

for carrying the radium,  $R$ ;  $J$  is a hollow cylinder having its 2½-in. wall filled with .02-in. shot;  $D$  is a brass tube (diam 6 in., length 6 in.) filled with solid lead through which is drilled an eccentrically placed orifice  $dd'$ , so that by rotation of  $D$  in its supporting saddle, the orifice can be either entirely open to, or completely shielded from the radiation from  $R$ ; and finally, the pressure tank with aluminum window,  $W$ , and ionization chamber,  $K$ .

The limiting aperture  $dd'$  was formed by first drilling a commensurate hole and then forcing through it a series of plug gages which gradually enlarged the opening and burnished it accurately to size. The carrying saddle was on ball bearings and provided with stops which insured accurate repetition of "open" and "closed" positions, 180° apart.

To establish proper alinement, the aperture  $dd'$  was first adjusted in height and direction so that the transmitted beam would pass

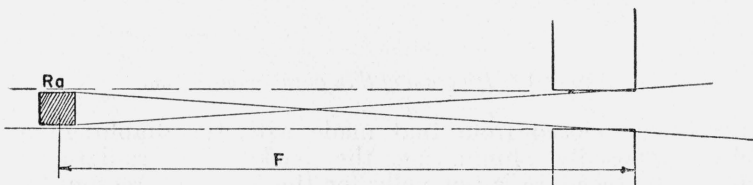


FIGURE 3.—Diagram showing the passage of radiation through the edges of the diaphragm.

centrally through the window,  $W$ , of the pressure tank and along the central axis of the ionization chamber. This adjustment was not at all critical. The plug gage finally used was then reinserted in  $dd'$ , fitting snugly and reaching through the radium carrier tube,  $cc'$ , which was then fixed in its position by filling the tank,  $A$ , with shot. The alinement of  $J$  simply required correct "open" and "closed" positions. On removing the plug gage, inserting the radium, and shifting the screen,  $N$ , into place, the set-up is ready for making measurements. The time required to effect this alinement does not exceed 15 minutes.

It is particularly important when dealing with highly penetrating radiations that in the operation of the shutters the stray radiation field is not changed. The rotary type of shutter here used fulfills this requirement. It was found that by using the collimating and screening system as described, the background radiation did not exceed 4 percent of that of the direct beam, and by inference, the error introduced by the quantity passing through the "plug" alone was negligible. This low background radiation was no doubt responsible for the low-shielding requirements around the electrometer system mentioned above.

With the type of collimating system here used, there will, of course, be a small amount of radiation transmitted through the edge of the diaphragm at the exit end of the orifice owing to the obliquity of the rays from the outer parts of the radium source (see fig. 3). The relative magnitude of this effect will decrease with increasing distance,  $F$ , between radium and diaphragm. Calculations were made for such leakage by dividing the exit face of the orifice into narrow concentric zones and then computing the transmission of each. The results given in table 1 are the maximum, and the actual transmission may possibly be less by as much as 10 percent. Since most of the other measurements were made at  $F=62.5$  cm, a 2-percent correction only was applied.

TABLE 1.—Transmission through edge of diaphragm in percentage of unobstructed radiation

$F$	Percent
<i>cm</i>	
50	2.7
60	2.2
65	1.8
70	1.7
75	1.6

Since the thickness of the diaphragm,  $dd'$ , is not small compared to its distance from the source of radiation, and since its wall is not perfectly opaque, the question arises whether or not the aperture at  $d'$  is, as for the ideal case, the limiting aperture for determining the quantity of ionization. For ideal conditions and all radiant points sufficiently close to the axis of the system, and sufficiently close to a fixed distance from the end,  $d'$ , the quantity of direct radiation passing through the diaphragm varies inversely as the square of the distance of the source from  $d'$ . The quantity of the radiation reaching the ionization chamber from the inner walls of the aperture is negligible, and that passing through the edges of the diaphragm is given in table 1. Of course, the radiation originates from an extended and not a point source; but as long as the diameter of the source is small compared to the distance,  $F$ , (1:70 in our case) and is smaller than the diaphragm diameter the inverse-square law may be expected to hold.

To test the question, ionization-chamber measurements on the transmitted radiation were made with the radium at a series of positions within its collimating tube,  $cc'$ ; that is, at different distances,  $F$ , between the radium and the exit face,  $d'$ , of the diaphragm. Corrections to these measurements were then made from table 1, and the results plotted against the inverse square of the distance—in figure 4. The plotted points fall very closely on a straight line through the origin and thus justify the assumption that the limiting aperture is at the exit face of the diaphragm; and also assure the practical correctness of the calculated magnitudes of table 1.

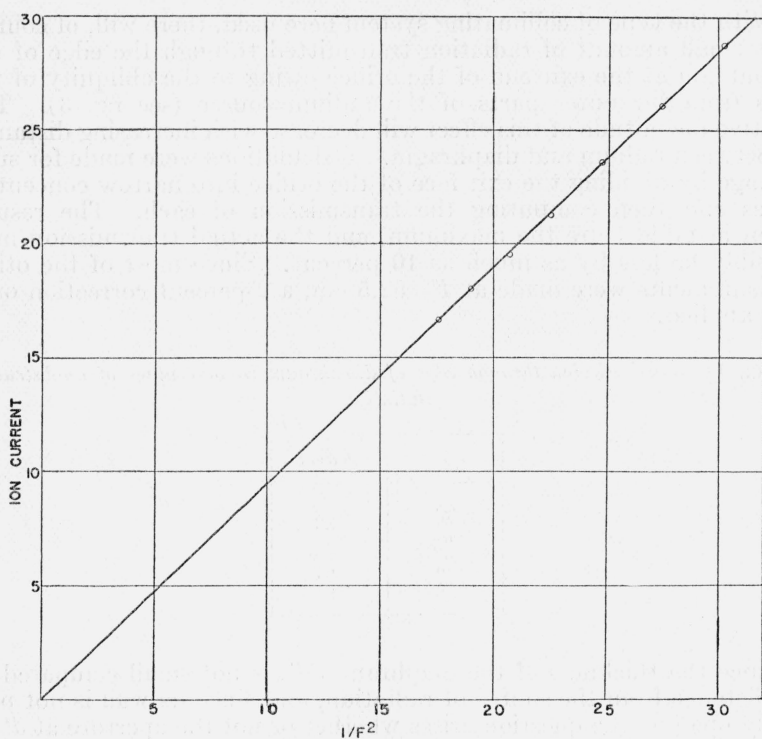


FIGURE 4.—Inverse-square-law plot for distance between radium and diaphragm edge.

#### IV. SATURATION

With the requisite plate spacing of about 30 cm and a pressure of 10 atm, saturation voltages are expected to be high, and in fact saturation is not completely obtainable because of columnar recombination at this pressure. Figure 5 shows a current-voltage curve of the ioniza-

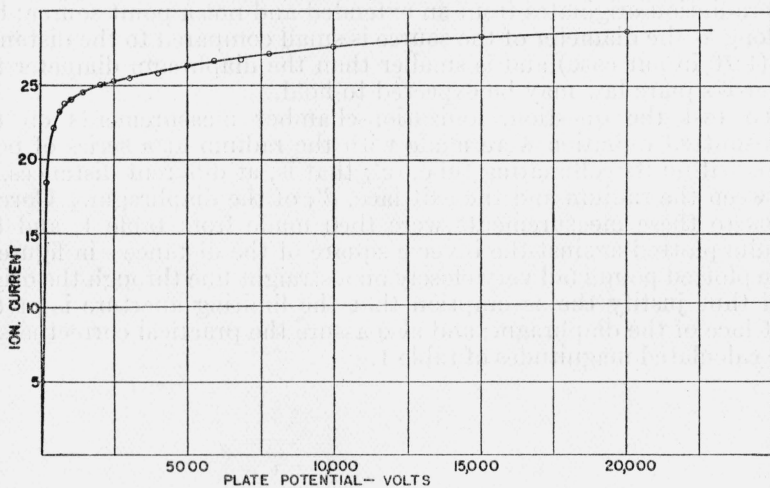


FIGURE 5.—Saturation curve at 10 atm.



tion chamber under such conditions. There is no evidence of saturation at the highest field strengths, 670 volt/cm, corresponding to the applied potential of  $2 \times 10^4$  volts. This condition is expected because it corresponds to only 67 volt/cm at 1 atm, which is barely sufficient to produce saturation at that pressure, even neglecting columnar recombination. Applied potentials above 20 kv could not, however, be used because the resulting unsteadiness exceeded the permissible limits.

The possibility of applying Jaffe's theory of columnar ionization for extrapolating to saturation conditions was suggested by our earlier work on the X-ray ionization of liquids [6, 7, 8], and later work by Zanstra [9] and by Clay and coworkers [10, 11], who have applied it to gases from 20 to 400 atm. Their findings have been further verified by Broxon and Meredith [4] with high-pressure cosmic-ray ionization chambers.

Zanstra gives the following relation between the current,  $i$ , and the saturation current,  $I$ :

$$\frac{1}{i} = \frac{1}{I} + \frac{q}{I} f(x), \quad (1)$$

where

$$\frac{q}{I} = \frac{\alpha N_0}{8DI} \quad (1a)$$

in which  $N_0$  is the number of ions per centimeter of path length,  $D$  is the diffusion coefficient, and  $\alpha$  is the recombination coefficient.  $f(x)$  is a Hankel cylinder function of the form

$$f(x) = e^x (j\pi/2) H_0^{(1)}(jx), \quad (2)$$

where

$$x = c \left( \frac{X}{p} \right)^2, \quad (3)$$

in which  $X$  is the field strength,  $p$  is the pressure in atmospheres,  $j = \sqrt{-1}$ , and for air,  $c = 1.24 \times 10^4$ .

Figure 6 gives a reproduction of Zanstra's curves  $f(x)$  versus  $X$  for the range of  $x=1$  to  $10^{-6}$ . A plot of  $1/i$  versus  $f(x)$  for different values of  $X$  should, by eq 1, give a straight line for which the intercept on the  $1/i$  axis is the reciprocal of the saturation current,  $I$ . It should be noted that the constants in eq 1 and the approximations in the theory do not directly influence the value of  $1/i$  but serve to rectify the curvature which would otherwise appear in a simple plot of  $1/i$  versus  $1/X$ .

The validity of the Jaffe-Zanstra theory was tested in the present work by measuring a fixed 300-kv X-ray beam at pressures of 1.0, 6.80 and 10.13 atm, respectively. In the ionization chamber, a plate separation of 30 cm was used, so that no wall correction was necessary at any pressure. Figure 7 shows the current-voltage curves at 1 and 10.13 atm (coordinate scales at right and top) and the  $1/i$  versus  $f(x)$  (scales at left and bottom) for 10.13 atm. The intercept of this  $1/i$  plot and a similar plot at 6.8 atm gave the following values of  $I$  after correction for air absorption:  $I$  (10.13 atm)/10.13 = 1.076;  $I$  (6.8 atm)/6.8 = 1.078; and  $I$  (1 atm) = 1.078. The agreement is better than the

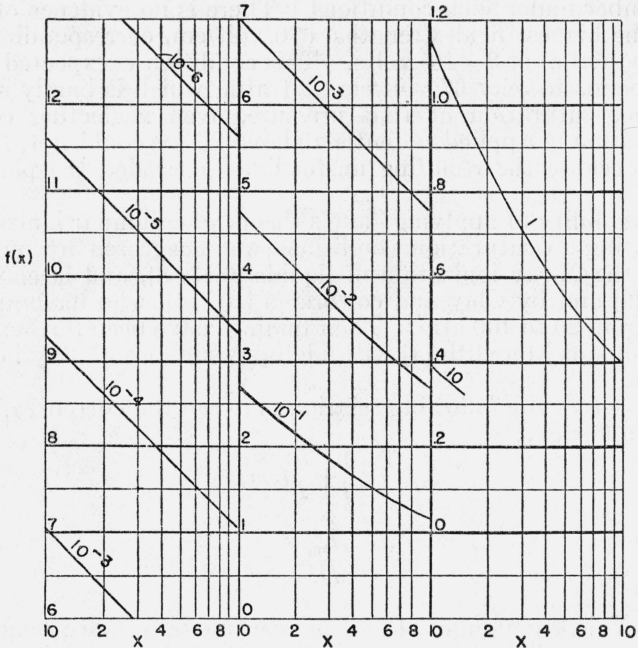


FIGURE 6.—Zanstra's curves for evaluation of  $f(x)$ .

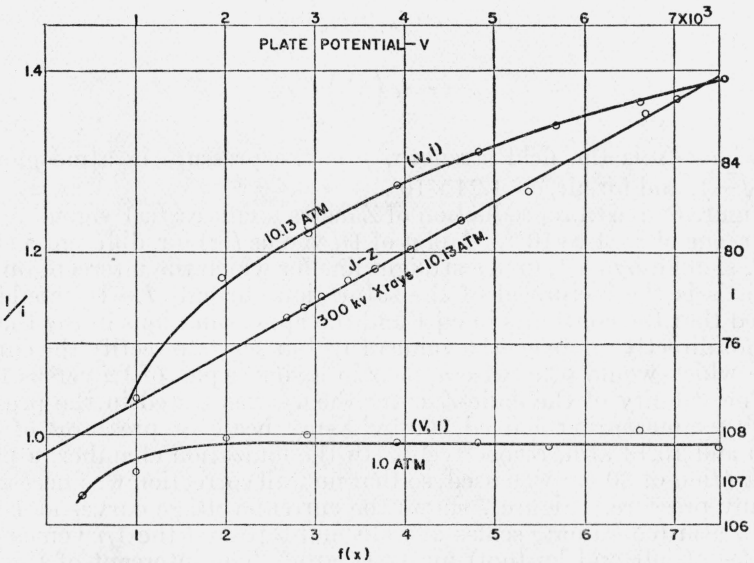


FIGURE 7.—Normal saturation and J-Z plots at 10 atm.

experimental error of about  $\pm 0.5$  percent in the particular measurements warrants. These measurements were made with a maximum of 8,000 volts on the plate (field strength = 27 v/cm.atm).

It was, of course, necessary to see if the measurements made at field strengths between 3.3 and 27 v/cm·atm formed an adequate basis for extrapolation or if higher fields were required. Figure 8 shows the  $1/i$  versus  $f(x)$  curve obtained from the data plotted in figure 5. Here it is evident that it is not necessary to exceed the field of 270 v/cm at 10 atm or 27 v/cm·atm, although by going to fields as high as 67 v/cm·atm the extrapolation is somewhat shortened. Under the particular conditions at 27 v/cm·atm the current is 87 percent of the saturation current,  $I$ , and at 67 v/cm·atm it is 94 percent.

The extrapolation, therefore, amounts here to about 6 percent of the total length, as compared with values as large as 45 percent for

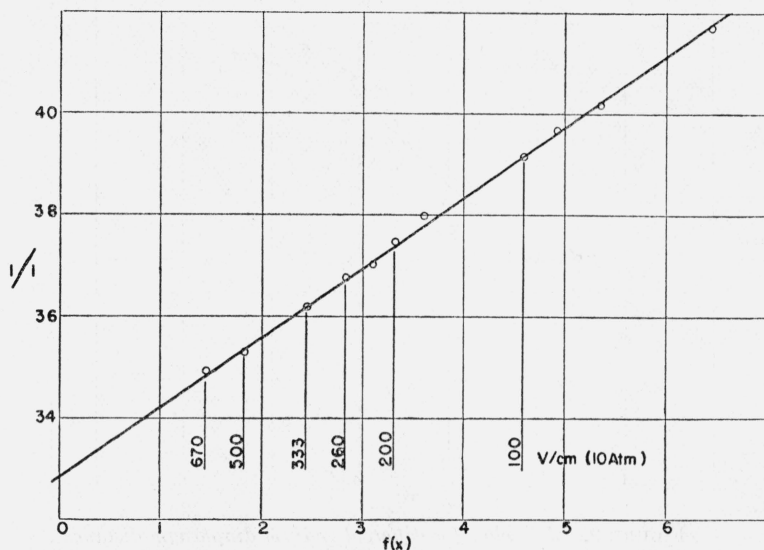


FIGURE 8.— $J$ - $Z$  plot for field strengths up to 670 v/cm.

higher pressures and the higher fields used by Clay [10 to 12] and Broxon and Meredith [4]. (Reduced to atmospheric pressure, their field strengths were usually much smaller than ours.) By decreasing the plate separation to 18 and 7 cm, the maximum field strength was increased to 45 and 80 v/cm·atm, respectively, and the maximum measured current was 96 and 97 percent, respectively, of the saturation value obtained by the  $J$ - $Z$  extrapolation.

The chamber was not usable for our purposes under these conditions, since the decreased plate separations would introduce undesirable wall effects. The results indicated, however, that the  $J$ - $Z$  plot would be linear to within at least 3 percent of the saturation current value and hence the extrapolation used with the wider plate separations is similarly justifiable.

For convenience it is frequently desirable to make comparative measurements without, for each condition, going through the entire  $J$ - $Z$  extrapolation procedure. Such a procedure is permissible under conditions (1) where the ionization density does not vary over too wide limits, or (2) where measurements are well inside the region

where the  $J$ - $Z$  curve is linear and the field strengths do not differ too greatly, or (3) both.

A check of the first point was afforded in testing the validity of the inverse-square-law plot of figure 4. The points on this plot were obtained at a fixed field strength of about  $26\text{v/cm}\cdot\text{atm}$ , giving a value for  $f(x)$  of about 3. To check their validity, complete  $J$ - $Z$  extrapolations (fig. 9) were made for two distances between the radium and the exit face of the diaphragm, such that the edge correction for the diaphragm should be practically negligible between the two positions. From the  $J$ - $Z$  computations the saturation value for 61.0 cm is 0.0273

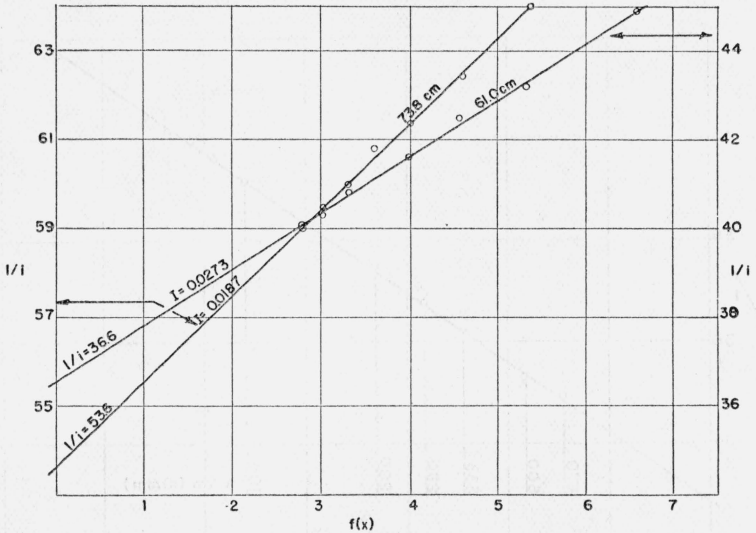


FIGURE 9.— $J$ - $Z$  plots for different radium-diaphragm distances.

and for 73.8 cm is 0.01865. The ratio  $0.0273/0.01865=1.464$  should, if the theory is applicable, be that of the ionization produced in the two cases, which in turn should be inversely proportional to the squares of the distance. This latter ratio is  $(73.8/61.1)^2=1.458$ , and agrees with the ratios of the ionization to 0.4 percent, which is within the experimental error. It is clear from the plotted points (fig. 9) that even with the poorest choice of the position of the intercept the saturation value could not be in error by as much as 0.5 percent.

The second point is illustrated by the curves in figure 10, where two  $J$ - $Z$  curves are drawn through points measured for the same beam (hence, same ionization density) but with different plate separations and hence, different field strengths. The resultant straight lines converge slightly, so that the ratio of the two ordinates for any single value of  $f(x)$  over the range of  $f(x)$  shown has the constant value 1.023. Similar ratios at a given applied voltage are very nearly 1.00. This divergence arises from the difference in field strengths and will be discussed later.

It should be pointed out that Zanstra's evaluation of Jaffe's equation fails for low field strengths, so that the  $J$ - $Z$  plot is no longer linear, the points falling above the extended straight line. The point

at which this deviation begins is variable and depends upon the nature of the gas, pressure, ion density, and field strength. Under the conditions of this study, the deviation begins for fields below about 2 vicm·atm or values of  $f(x) > 7$ . The effect has been described by Clay and van Kleef as the condition where volume recombination begins to become relatively dominant in comparison with the columnar recombination.

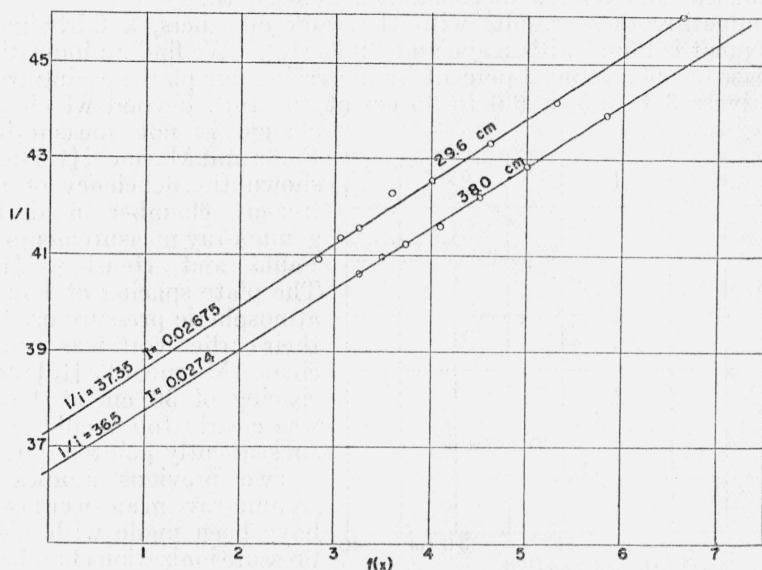


FIGURE 10.— $J$ - $Z$  plots for different plate separations.

## V. EFFECT OF PLATE SEPARATION

In their gamma-ray measurements with a large open-air guarded-field ionization chamber, Kaye and Binks [13] found a plate separation of about 3.0 m apparently sufficient to prevent the impingement of an appreciable amount of secondary radiation upon the plates. On this basis we have done much of our exploratory work with a separation of 30 cm at a pressure of 10 atm, which is equivalent to Kaye's spacing.

However, to be certain that the chamber was free from wall effects, ionization currents were measured for different plate separations. For these, the beam width was about 2 cm at the center of the chamber. Curve *A* in figure 11 was obtained by simply increasing the plate spacing while keeping a fixed potential of 7,000 volts between plates. Saturation was apparently reached at about 30 cm. However, with increasing distance between the plates, there is a proportionate decrease in field strength; consequently, the flattening of the curve, *A*, may be caused in part by the decrease in field strength. To eliminate this factor, the second curve, *B*, was obtained with the same field strength at all plate spacings. In this latter case, field saturation was not definitely reached below about 35 cm, which spacing, or one greater, was used in the final radiation measurements. These studies have shown the unreliability of comparing ionization

measurements made under different field strengths. Above about 2.00 v/cm·atm, comparisons at the same field strength are valid, provided the radiation intensities and qualities are about the same.

The results of curve *B* in figure 11 were checked by means of the *J-Z* extrapolation made at plate spacings of 29.6 and 38.0 cm and shown in figure 10. The ratio  $I_{38}/I_{29.6}=0.02740/0.02675$  is 1.023 as compared with a ratio  $i_{38}/i_{29.6}=26.8/26.3$  which is 1.019 (from figure 11) for the curves made at constant field strength.

Comparing these results with the work of others, a fairly good agreement is found with Kaye and Binks [13]. We find an ionization increase of only about 2 percent by increasing our plate spacing from effectively 3 to 3.5 m (30 to 35 cm at 10 atm), beyond which the change is not measurable.

Faila and Marinelli [14] have shown the deficiency of the free-air chamber used for gamma-ray measurements by Faila and Henshaw [15]. The plate spacing of 1 m at atmospheric pressure used in their earlier work was insufficient. Mayneord's [16] plate spacing of 30 cm at 1 atm was clearly too small, as he subsequently pointed out.

Two previous studies of gamma-ray measurement have been made with high-pressure ionization chambers. Clay and van Kleef [17] used an argon-filled chamber with metal-grid electrodes 5 mm apart, at fields from 100 to 4,000 v/cm at 100-atm pressure. (This was equivalent to 40 v/cm·atm). Clay and van Tijn [18] obtained with this chamber, by means of the *J-Z* extrapolation, a value for Eve's constant which is in reasonable agreement with other determinations. However, since their plates had an effective separation of only about 50 cm at 1 atm, wall effects played some part in their result.

Friedrich, Schulze, and Henschke [19] used air at pressures up to 139 atm in a graphite chamber having an electrode separation of what appears to be 8 mm in their drawing; but they took no account of columnar ionization. Their effective plate separation at 1 atm was on this basis about 110 cm, hence the measurements also involve wall effect.

## VI. ELECTRONIC EQUILIBRIUM

To insure the accurate evaluation, in roentgens, of gamma rays, using a free-air chamber, it is necessary to have what is known as a state of electronic equilibrium over the region from which the ions are measured. This condition is realized when the electrons which escape from the measuring volume are exactly compensated for by electrons which enter it from the outside space. Applied to the

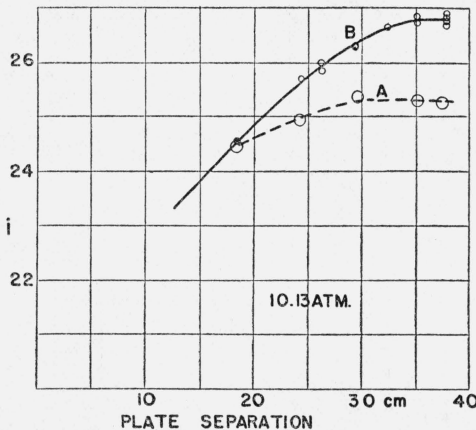


FIGURE 11.—Plate separation curves at 10.13 atm.

measurement of gamma radiation from radium by the free-air ionization chamber, this requires that the volume from which the ions are measured be surrounded by an envelope of air having a thickness in all directions at least equal to the maximum range of the secondary electrons in those directions.

Where a narrow collimated beam of radiation is involved, this requires that between any limiting diaphragm or filter and the ionization chamber, there must be a free-air path of a length at least equal to the longest paths of any scattered or recoil electrons. Since the secondary electrons involved in gamma-ray scattering are predominantly in the forward direction, a very considerable distance between the lead diaphragm,  $dd'$ , and the collector electrodes of the ionization chamber is required. Kaye and Binks [13] found that at atmospheric pressure a distance of 7 m was apparently sufficient, whereas Friedrich's measurements [19] indicated about 15. Using our equipment at 10 atm,

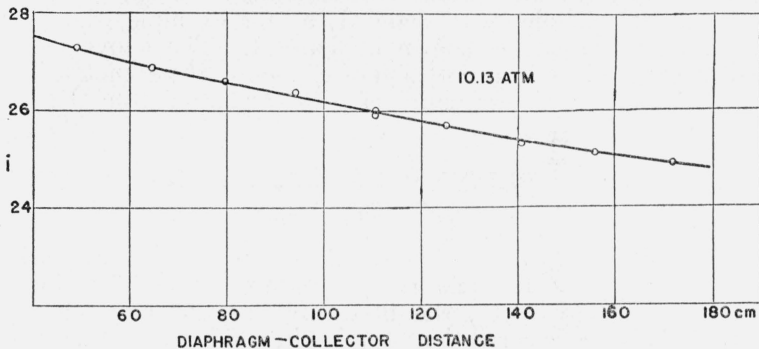


FIGURE 12.—Air absorption of gamma rays at 10 atm.

it was possible to test this over a range equivalent to 6 to 21 m at atmospheric pressure.

Following our earlier procedures [1], ionization measurements were made for distances between the collector and the window over the above range. Inasmuch as the diaphragm was but 10 cm outside the aluminum window of the pressure tank, distances from the aluminum window to the collector were used. Figure 12 shows the results of these measurements. It is seen that the decrease in ionization is linear within less than 0.5 percent over the range of 70 to 150 cm, which is equivalent to a range of 7 to 15 m at 1 atm. The small increase, above 150 cm and below 70 cm, may be ascribed to scattering from the ends of the pressure cylinder. All measurements given in this paper were made at distances equivalent to 9 m at 1 atm unless otherwise noted.

It is clearly evident from the measurements in figure 12 that a state of electronic equilibrium existed in the forward direction. From plate-separation measurements of figure 11, curve *B*, a similar state in the lateral direction is assured. However, a further check on these conclusions was obtained in trying to find Failla's "cloud effect" [14]. Two sets of measurements were made at distances,  $L$ , between the collector and the window of 58.9 and 120.1 cm, wherein the satura-

tion currents were determined by the *J-Z* method. The ratio of the corresponding values of *I* was 1.023, as compared with a ratio of 1.027 derived from figure 12 for the air absorption alone.

These measurements show the existence of electronic equilibrium within experimental errors but does not provide a fair test for the cloud effect. Since our minimum distance to the limiting diaphragm was about 6 m, we may have been outside the region where any expansion of the cloud could be detected. Also, our collector having an effective length of 2.5 m at 1 atm, any *small* change in the effect of the cloud shape would be masked. Any attempt to verify the effect at lower pressure would have been unreliable, because then a state of lateral electronic equilibrium would no longer exist.

As indicated, the data in figure 12 may be used to determine the air absorption for the radiation used. The absorption, which is found to be 7.7 percent per meter at 10 atm, requires that a correction be applied to all absolute measurements or all comparative measurements wherein *L* is changed; as for example, the inverse-square-law measurements shown in figure 4. The above value reduced to 1 atm gives an absorption of 0.77 percent per meter, in good agreement with the lower figure of 0.75 calculated from Kaye and Binks' work [13].

## VII. THE MEASUREMENT OF GAMMA RADIATION IN ROENTGENS

The magnitude of gamma radiation in roentgens has been variously defined in terms of the quantity of radium, time, filtration, and distance. In many cases, the literature has not been clear with regard to the various corrections applied in making the measurement, and this has resulted in confusion in the final results. We shall, therefore, risking repetition, describe in detail the various measurements and corrections employed in this study.

For reference purposes, the "emission constant,"  $Q_0$  of radium is defined as the number of roentgens measured in 1 second at 1 cm from 1 g of radium, assuming the complete exclusion of any beta rays. This implies that the radium salt must be enclosed in a filtering container of known gamma-ray absorption or by an air envelope of equivalent thickness.

In the radiological field, however, the quantity of gamma radiation,  $Q$ , from radium is expressed as equal to the number of roentgens per hour produced by each milligram of radium, acting through 0.5 mm of platinum, at a distance of 1 cm from the radium. Since the radiation varies inversely as the square of the distance, the gamma radiation is then

$$Q = r/D^2 m T,$$

where  $r$  is the number of roentgens at a distance of  $D$  centimeters from  $m$  milligrams of radium, in  $T$  hours. Here, the quantity of radium is taken as the actual mass of radium  $B$  and  $C$  in equilibrium with their disintegration products. The 0.5-mm platinum filtration is added for the principal purpose of eliminating any beta radiation, and enters the experiment as a purely arbitrary factor for which no absorption correction is made. (Should correction be made for the absorption of the gamma radiation in the platinum, the quantity



result would be several percent larger, and not expressed consistently with the radiological definition.)

The radium employed in this study was of the order of 500 mg contained in a platinum cylinder 4.45 mm long and 9.95 mm outside diameter, having walls 0.500 mm thick. It was necessary to know the actual radium content for the proper evaluation of our results. The method of calibration is that uniformly used in the national laboratories of the various countries and has been recently described by Perry [23].

Our radium was calibrated in the National Bureau of Standards radium laboratory under the supervision of L. F. Curtiss. The usual substitution method was employed, and he states that the measurements are believed to be accurate within 0.1 percent. The electroscope was encased in a thick lead shield and, hence, differed markedly from our free-air ionization chamber. The comparison was made against a number of individually calibrated radium preparations contained in thin-glass tubes and bunched together so as to approach as nearly as possible the geometrical configuration of the preparation under calibration. By this means the absorption of radiation in the salt of the unknown and the comparison sources was made as nearly the same as possible and, therefore, balanced out. To test for this, the comparison tubes were arranged in a variety of reasonable configurations, and it was found that any calibration differences were negligible. The final result would not, of course, give the actual number of milligrams in the unknown, since the radiation from the latter was filtered by the additional 0.5 mm of platinum, while that from the standard was measured after passing through the glass only.

The Bureau's certificate stated that the sample had a gamma radiation equivalent to that from 466.6 mg of radium.<sup>4</sup> Correction must be made, however, for the absorption of the emitted radiation in the salt itself (radium sulfate), because the amount of radiation finally emerging varies with the configuration of the salt. An absorption coefficient applicable to this problem was very kindly supplied to us by G. C. Laurence.<sup>5</sup> He gives a value of 0.037 cm<sup>2</sup>/g, which he determined under experimental conditions comparable to ours. For making this test, he placed the radium salt in a funnel-shaped thin-walled tube. In one measurement the radium was spread thinly over a considerable area normal to the direction of measurement, and hence offered a negligible absorption of the radiation in the salt. In the second measurement, the container was inverted, so that the salt was packed in a long thin stem such that in its lengthwise direction a considerable absorption of the radiation took place. The absorption coefficient was then derived from the difference between the two measured radiations.<sup>6</sup>

Assuming that, on the average, all the radiation is absorbed by only half of the radium content, an absorption value of 1.47 percent is derived. This is in agreement with an integrated value, since it must

<sup>4</sup> Had the radium been contained in a thin-glass tube (as is used for the standards) its gamma radiation would have been 4.3 percent greater than the certified value. (Under the conditions of calibration with a lead-walled electroscope, the platinum absorbed 4.3 percent of the radiation.) Therefore, the *actual radium-element content* of the preparation here used was 486.6 mg.

<sup>5</sup> National Research Council of Canada. Personal communication.

<sup>6</sup> Laurence points out that absorption of the gamma rays in the source itself is complicated by the fact that we are dealing with the net absorption, which is a difference between the true total absorption and scattered radiation. The net-absorption coefficient will be considerably lower than the more usually quoted coefficients, which are total true absorption. Since the net absorption is so small, the effect on the absorption coefficient of any change in radiation quality is negligible.

be assumed that there is no change in the quality of the radiation in traversing the salt—a not unreasonable assumption since the total absorption is so small.

Another absorption figure was supplied to us by Dr. G. W. C. Kaye.<sup>7</sup> For a cylinder of approximately the same diameter as ours but with 2.5 mm inside length, he has determined a correction of 1.6 percent, which for our conditions would amount to nearly 3 percent—a figure higher than any of those computed above. His work is expected to be published at an early date.

In the final calculations a correction of 1.47 percent for absorption in the salt has been applied to the radiation from 486.6 mg *actual* content of the salt of our capsule. We thus deal with an *effective radium content* of 479.4 mg in the final result.

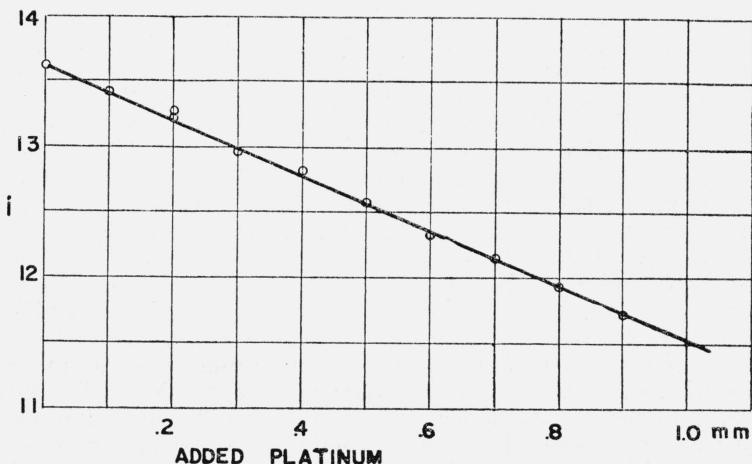


FIGURE 13.—Platinum absorption of gamma rays.

Although not very great, the correction for the absorption of the gamma radiation in the radium salt probably represents the largest uncertainty in our experimental results.

It should be emphasized that the 4.3-percent platinum-absorption correction in the original radium calibration applies only to the particular measurement technique employed by the radium laboratory. To make possible correction of our results to other platinum filtrations, separate platinum-absorption measurements were made using the measurement technique described in previous sections. Successive layers of 0.10-mm platinum foil were cemented lightly to the face of the capsule and ionization readings made with the back of the capsule always in the same position within the collimating tube (figure 2). The results, shown in figure 13, indicate uniform absorption over the range of 1 mm of added platinum. The linearity of the absorption curve would, therefore, indicate that under our conditions of measurement, the 0.5-mm platinum wall absorbs approxi-

<sup>7</sup> National Physical Laboratory. Personal communication.

mately 7.2 percent of the gamma radiation. This figure does not enter our calculations of  $Q$ , since the filtration by 0.5 mm of platinum is included in the definition, but does enter in the evaluation of the emission constant,  $Q_0$ .

To obtain an estimate of the precision of the measurement, the errors in the several quantities involved in the determination of the emission constant of radium and the degree to which they enter the final results are listed in table 2.

TABLE 2.—Errors involved in the measurement of gamma rays

Quantity	Residual error	Error in result
<i>Group A:</i>	<i>Percent</i>	<i>Percent</i>
Radium calibration.....	$\pm 0.1$	$\pm 0.1$
Salt absorption.....	20	.4
<i>Group B:</i>		
Air absorption.....	.3	.2
Diaphragm edge.....	10	.2
Distance of radium.....	0.2	-----
Current measurement.....	.3	.3
Saturation current.....	.3	.3
Volume measurement.....	.2	.2
Pressure.....	.3	.3
Average deviation from mean in all observations.....	.5	-----

The errors fall into two groups: *A*, those involved in determining the effective quantity of radium; and *B*, those involved in measuring the gamma radiation by our set-up.

In group *A* the calibration of the radium is accepted with the estimated possible error. Likewise, the residual error in the absorption by the salt lies in the uncertainty of Laurence's absorption coefficient.<sup>8</sup>

In group *B*, also, the residual errors in each measurement are as indicated. The average deviation from the mean in all observations did not exceed 0.5 percent. The possible error in distance of the radium enters, because this is measured to the center of the capsule, whereas it might be argued that the distance should be measured to some other point. The residual error in deriving the saturation current is obtained from the greatest possible spread in the value of  $1/I$  for  $f(x)=0$  due to the choice of the straight line drawn through the plotted points.

The accuracy of measurement obtained by the square root of the sums of the squares of the individual errors in group *B* alone is  $\pm 0.25$  percent and for both groups, *A* and *B* together,  $\pm 0.28$  percent. Since the sum of the errors in group *A* is 0.41 percent, it is seen that the effect of the errors of measurement in the present work on the final result is negligible in comparison with the effect of the uncertainty in the quantity of radium and loss by self-absorption.

In table 3 are given the various experimental values and apparatus constants involved in the measurement of gamma rays in roentgens.

<sup>8</sup> Since a question of interpretation is involved in this correction, there is a possibility of as much as a 2-percent error in the final result, depending upon the particular coefficient used. The use of any other probable coefficients will tend to increase the final value of the emission constant.

TABLE 3.—Factors in the calculations of the emission constant

Capacitance ( <i>C</i> ).....	851 $\mu\text{f}$
Compensating volts/sec ( <i>V/i</i> ).....	$1.791 \times 10^{-2}$ (extrapolated value)
Distance ( <i>D</i> ).....	62.0 cm
Pressure ( <i>P</i> ).....	9.67 atm
Temperature ( <i>T</i> ).....	302° K
Measuring volume ( <i>W</i> ).....	19.60 cm <sup>3</sup>
Platinum absorption factor ( <i>k<sub>p</sub></i> ).....	1.072
Effective radium content ( <i>R<sub>e</sub></i> ).....	479.4 mg
Air-absorption factor ( <i>k<sub>a</sub></i> ).....	1.081
Slit edge-correction factor ( <i>k<sub>s</sub></i> ).....	0.980

The following equation will give the emission constant,  $Q_0$ , for radium *B* and *C* after allowing 7.2 percent for the absorption of the radiation in platinum:

$$Q_0 = \frac{0.9 \times CV \times T \times D^2 \times k_p k_a k_s}{300 \times W \times t \times 273 \times R_e \times P} \quad (5)$$

$$= 2.43 \pm 0.01 r/g \cdot \text{sec at 1 cm.}$$

In terms of the radiological unit the emission is

$$Q = 8.16 \pm 0.04 r/mg \cdot \text{hr at 1 cm.}$$

Various summaries have been made of the experimental values obtained for  $Q$  using thimble and other chambers, which, for comparison with our results, we give in table 4. In this table, column 3 gives the number of separate previous determinations of  $Q$ , which are averaged in column 4. Column 5 gives the maximum divergence between the averaged values. Column 6 gives the new value presented by each author.

TABLE 4.—Summary of thimble chamber measurements

Summary by—	Reference	Number of determinations	Average value, $Q$	Spread	Author's own value
				<i>Percent</i>	
Mayneord (1934).....	[20]	5	8.6	13	8.3
Kaye and Binks (1938).....	[13]	9	8.0	17	<sup>a</sup> 8.0
Friedrich (1938).....	[19]	9	8.15	26	7.8
Rovner (1938).....	[21]	12	8.55	15	-----
Laurence (1938).....	[22]	6	8.35	9	-----

<sup>a</sup> Free-air chamber as well as thimble chamber.

It is interesting to note that while the above summaries are based on essentially the same experimental sources, they vary over a range of some 7 percent. This is because the different workers, in preparing the summaries, have had to correct the final results of the other workers for filtration or salt absorption and have used, for the purpose, differing correction factors. Other observers have weighted

their summaries by discarding results which for one reason or another seemed questionable to them. This is, of course, a perfectly valid procedure, but it does show that the most probable value of  $Q$  is not to be had by a simple averaging of the various final results obtained in many different laboratories.

Our own value of  $Q=8.16$  r/mg-hr at 1 cm is about 2 percent higher than the only other value obtained under free-air conditions—8.0 r/mg-hr at 1 cm by Kaye and Binks. However, if we had used their value for the salt absorption, our result would have been still higher by about  $1\frac{1}{2}$  percent, and hence the disparity even greater.

Our own final values of  $Q_0$  and  $Q$  are presented with reservations as to the accuracy of the radium-salt absorption correction, and further corrections may be made in the future. Further studies by experts in the radium field may yield a more accurate evaluation of this absorption coefficient.

### VIII. CONCLUSION

The primary purpose of this investigation has been to investigate the possibilities of measuring the very short wavelength radiation by means of a pressure ionization chamber. The results with gamma rays show that such wavelengths may be satisfactorily measured in roentgens so long as the pressure is sufficient to give an effective free path between the beam and measuring electrodes equal to the longest range of the recoil electrons. Since the ranges overlap those involved in X-ray scattering up to 1.5 MV, it may be concluded that X-rays up to at least 1.5 MV may be measured in roentgens under free-air conditions. It is, therefore, possible to directly calibrate thimble chambers against a free-air standard for all excitations presently available, and thus provide a logical extension of the useful range of thimble chambers. Likewise, having once established a value for r/mg-hr at 1 cm, it is possible to effect a direct calibration of radioactive preparations in terms of roentgens without further reference to the primary radium-salt standards.

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WASHINGTON, January 20, 1940.